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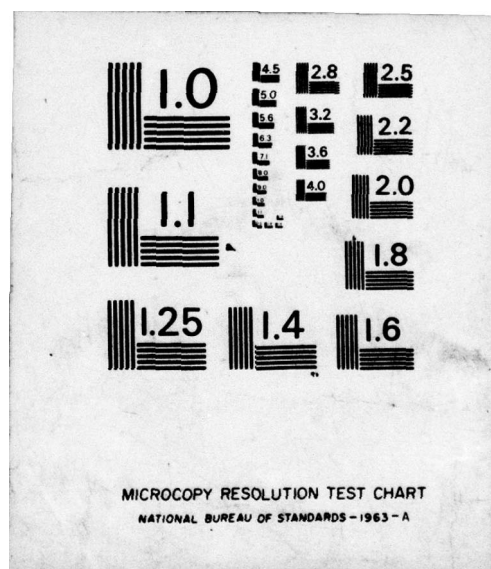
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TECHNICAL REPORT

WHITE OAK LABORATORY

MAGNETOSTRICTION AND STRUCTURAL DISTORTION IN RARE EARTH INTERMETALLICS

BY
James R. Cullen
Arthur E. Clark

1 NOVEMBER 1976

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| REPORT DOCUMENTATION PAGE | | READ INSTRUCTIONS BEFORE COMPLETING FORM |
|---|--|---|
| 1. REPORT NUMBER | 2. GOVT ACCESSION NO. | 3. RECIPIENT'S CATALOG NUMBER |
| 14 NSWC/WOL/TR-75-177 | | |
| 4. TITLE (and Subtitle) | 5. TYPE OF REPORT & PERIOD COVERED | |
| 6 Magnetostriction and Structural Distortion in Rare Earth Intermetallics | | |
| 7. AUTHOR(s) | 8. CONTRACT OR GRANT NUMBER(s) | |
| 10 James R./Cullen Arthur E./Clark | | |
| 9. PERFORMING ORGANIZATION NAME AND ADDRESS | 10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS | |
| Naval Surface Weapons Center White Oak Laboratory White Oak, Silver Spring, Maryland 20910 | 61153N; RR02-206; RR02-206-01; WR3811 | |
| 11. CONTROLLING OFFICE NAME AND ADDRESS | 11. REPORT DATE | 12. REPORT DATE |
| | 1 November 1976 | |
| | 13. NUMBER OF PAGES | |
| | 16 | |
| 14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office) | 15. SECURITY CLASS. (of this report) | |
| 12 18p | UNCLASSIFIED | |
| 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE | | |
| 16. DISTRIBUTION STATEMENT (of this Report) | | |
| Approved for public release; distribution unlimited | | |
| 16 RR02206 17 RR0220601 | | |
| 17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report) | | |
| 18. SUPPLEMENTARY NOTES | | |
| 19. KEY WORDS (Continue on reverse side if necessary and identify by block number) | | |
| Magnetostrictive Materials, Elastic Constants, Internal Distortion, Magnetic Anisotropy. | | |
| 20. ABSTRACT (Continue on reverse side if necessary and identify by block number) | | |
| On the basis of the special symmetry of the rare earth site in the Laves phase cubic compounds RT_2 (R = rare earth, T = Fe, Co, Ni) we predict the occurrence of a structural distortion which vanishes only when $[100]$ is the easy magnetization direction. An extraordinary external strain accompanies the distortion and provides an explanation of the observed size and anisotropy of the magnetostriction in RFe_2 . | | |

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Magnetostriction and Structural Distortion In Rare Earth Intermetallics.

The research reported herein was carried out in the Solid State Branch of the Materials Division of the Research and Technology Department under the ONR sponsorship (Task No. NOL-637/RR02-206-01). The work was also supported in part by the Naval Ship Systems Command and the Naval Surface Weapons Center Independent Research Program.

J. R. Dixon
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INTRODUCTION

An often studied and important series of rare earth-transition metal compounds are those that form in the cubic Laves or C15 phase. In this structure there are two rare earth and four transition metal atoms in the unit cell. The rare earth atoms have tetrahedral (T_d) site symmetry, i.e., the positions of the carbon atoms in the diamond structure, while the lighter metal atoms have D_{3d} symmetry. (See Figure 1.) In the compounds with Fe, the rare earth spins are taken to be parallel to one another and antiparallel to the iron spins, a picture which is consistent with all relevant experiments.⁽¹⁾ Rare earth compounds with iron in the Laves (C15) phase are strongly magnetic to well above room temperature, and $TbFe_2$ has the largest known magnetostriction and magnetic anisotropy at room temperature.⁽²⁾ These last two properties derive from the rare earth, and accordingly disappear for those magnetic compounds like YFe_2 and $GdFe_2$, where the rare earth atom has no orbital moment. It has also become increasingly clear⁽³⁾ that the magnetostriction for these compounds is very anisotropic. For example, $TbFe_2$ which has its easy magnetization direction parallel [111] develops strains of 2×10^{-3} , while $DyFe_2$, where [100] is easy exhibits strains of 10^{-5} or less. Since rare earth environments in these compounds are identical in every way, this anisotropy is expected to be a common feature of all the compounds. Thus we take as a fact that $\lambda_{111} \gg \lambda_{100}$ for Laves phase rare earth intermetallics. This is unusual for cubic metals. The cubic ferromagnets Fe and Ni for instance, show no such effect. We expect little anisotropy in the elastic constants. If the magnetoelastic coupling energies B_1 (100 easy) and B_2 (111 easy) are the same, the predicted⁽⁴⁾ ratio of strain in $TbFe_2$ to that in $DyFe_2$ is about two; the experiments give a ratio more like one hundred.

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- (1) Taylor, K. N. R., *Advances in Physics*, 20, 551 (1971). This article gives an extensive review of pertinent experiments relating to rare earth intermetallics.
- (2) Clark, A. E. and Belson, H. S., 17th Conference on Magnetism and Magnetic Materials, Chicago, Illinois, AIP Conference Proceedings No. 5, 1498 (1972); (*Phys. Rev. B* 5, 3642 (1972)) Clark, A., Cullen, J., and Sato, K., *Proc. 20th Conf. on Magnetism and Magnetic Materials*, AIP Conf. Proc. 24, 670 (1975).
- (3) Clark, A. E., 19th Conference on Magnetism and Magnetic Materials Nov 1973, AIP Conference Proc. 18, 1015 (1974)
- (4) The prediction is based on the use of equations (9) and (12) of this paper.

In this paper, we describe a theory for the spontaneous distortions of the atoms in the cubic Laves phase lattice due to coupling of the crystalline electrostatic fields with the total angular momentum of the 4f electrons.⁽⁵⁾ In the usual picture of magnetostriction, this distortion is a uniform strain in either of the two external strain modes: $2\epsilon_{zz} - \epsilon_{xx} - \epsilon_{yy}$, $\epsilon_{xx} - \epsilon_{yy}$, i.e., a distortion along 100 (λ_{100}), and ϵ_{xy} , ϵ_{yz} , ϵ_{xz} , a distortion along 111 (λ_{111}) where x, y, z refer to a cartesian set of axes parallel to the [100] directions. We show here that in the Cl5 structure, where there are two rare earth atoms in the unit cell, the crystalline field-angular momentum coupling gives rise to an internal distortion which in turn is coupled to an external strain.⁽⁶⁾ The results are a predicted internal distortion of the rare earth sublattice and an additional, extra-ordinary contribution to the external strain. This additional strain offers a natural explanation for both the size and anisotropy of the magnetostriction in the Laves phase intermetallics.

In the succeeding sections of this paper we give the physical picture of the process by which a Cl5 crystal distorts in the presence of a net magnetic moment. We then calculate the magnitudes of the distortions as a function of crystal field energies, internal and external moduli, and the shape of the 4f electron charge distribution. We give numerical estimates of the sizes of the predicted effects based on a point-charge model and experimental values for the moduli.

1. The Physical Picture

The way in which a spontaneous distortion occurs can be seen as follows: Suppose the magnetic moment is parallel to a [111] direction. Because of strong spin-orbit coupling the charge density of every rare earth atom takes on a very special shape for 4f electrons. For Tb, for instance, this density is pancake-like, (oblate) lying in a plane perpendicular to the [111] direction, as shown in Figure 2. The charge cloud of the atom labelled A in this figure comes closer to the three near neighbor atoms labelled B' than to its near neighbor at B. Because of a net coulomb attraction of this charge with the positive cores of its neighbors, the A atom moves toward the B' atoms, i.e., down in the Figure. Exactly the

- (5) The Phenomenology of Magnetostriction was first presented in R. Becker and W. Döring, *Ferromagnetismus* (Verlag Julius Springer, Berlin, (1939) P. 132, 145. See also C. Kittel, "Introduction to Solid State Physics" 2nd edition John Wiley and Sons, Inc. (New York). Reference 9 of this paper describes a microscopic calculation of magnetostriction of rare earth metals.
- (6) The possibility of internal distortions coupled to external strain is mentioned in E. R. Callen and H. B. Callen, *Phys. Rev.* **129**, 578 (1963).

opposite occurs at B; this atom moves up in response to the net attraction of A' atoms over the A atom. Figure 3 shows the situation when the magnetization is along [001], the easy axis for DyFe₂ and HoFe₂. The charge cloud remains equidistant from all its neighbors, and no distortion can occur. We now describe the calculation of this distortion and the consequent external strain.

2. Calculation of the Strains

The Electron-Lattice Interaction - Consider a 4f electron associated with a rare earth atom at the origin. Its wave function is changed from its atomic form by the crystalline potential of the nearby rare earth and iron atoms. The electron is described by its total angular momentum vector J. An effective Hamiltonian for a given site can be derived for the crystalline field by replacing the electron coordinates by their angular momentum operator equivalents.⁽⁷⁾ The first step is to obtain the correct form of the crystalline field.

For a site with tetrahedral symmetry,

$$V_{cf} = v_2 (x^2 + y^2 + z^2) + v_3 xyz + v_4 (x^4 + y^4 + z^4) + \dots$$

x, y, z are the dimensionless electron coordinates normalized by the lattice parameter, a_0 . Now because of parity, the average of the second term of V_{cf} vanishes; i.e., there is no effect from the odd-order term. Suppose, however, that the ion moves to a new position \underline{u}_A with coordinates u_{xA} , u_{yA} , u_{zA} say, with no change in size or shape of the unit cell. The energy of the 4f electron then depends on \underline{u} . To calculate $V_{cf}(\underline{u})$ we proceed as follows: In the electron-ion potential the electron coordinates x, y, z are replaced by $x - u_x$, $y - u_y$, $z - u_z$. We must also take into account the movement of the neighboring rare earth sublattice. Thus the ion coordinates X_n, Y_n, Z_n are replaced by $X_n - u_{xn}$, $Y_n - u_{yn}$, $Z_n - u_{zn}$. At this stage we make the assumption that $\underline{u}_n = \underline{u}_B$ for all near neighbors to an A site. Essentially, the reason this simplification works is that the near neighbors of a given rare earth on one sublattice (A) are all members of the other sublattice (B). Since the electron-ion potential contains x and x_n only in the form $x - x_n$, the distortion coordinates appears only as the difference in distortions on A and B sites. Define these coordinates to be u_x, u_y, u_z . We therefore have

$$V_{cf} = v_2^A \left[(x - u_x)^2 + (y - u_y)^2 + (z - u_z)^2 \right] + v_3^A (x - u_x) (y - u_y) (z - u_z)$$

⁽⁷⁾ Stevens, K. W. H., Proc. Phys. Soc. (London) 65, 209 (1952)

plus terms of higher order in $x-u_x$. The coefficients v_2, v_3 , etc., are lattice sums; in view of the special importance of the third order term it may be worthwhile giving the expression for v_3 on the point-charge model:

$$v_3^A = -15Ze^2 \sum_n \frac{x_n y_n z_n}{R_n^7} a_o^3 \quad 1(a)$$

Here Z is the effective number of charges on a rare earth ion and e is the electron charge. The sum is over all near neighbors. Now consider atom B at $(1/4, 1/4, 1/4)$. The form of the crystal field is the same, but u appears with opposite sign

$$V_{cf}^B = v_2^B [(x+u_x)^2 + (y+u_y)^2 + (z+u_z)^2] + v_3^B (x+u_x)(y+u_y)(z+u_z) + \dots$$

The different environments on A and B atoms have no effect on v_2 ; $v_2^A = v_2^B$. For the third-order term however, $v_3^A = -v_3^B$ as can be seen by examining the point charge expression for v_3 . For an A atom at the origin, the sum is over neighboring rare earths at $1/4, 1/4, 1/4$; $1/4, -1/4, -1/4$; $-1/4, -1/4, 1/4$; $-1/4, 1/4, -1/4$, in units of the lattice parameter. For a B atom (considered similarly located), the corresponding A neighbors would be at $-1/4, -1/4, -1/4$; $-1/4, 1/4, 1/4$; $1/4, 1/4, -1/4$; $1/4, -1/4, 1/4$.

So far, we have only considered a possible energy lowering by internal distortion, u . We can also expect that the symmetry change due to this distortion may drive an external strain as well, i.e., an extraordinary magnetostriction may appear, causing a shape change of the unit cell. We can take account of external strain and thereby account for both ordinary and extraordinary magnetostriction by writing:

$$x_n = x_n^o (1 + \epsilon_{xx}) + y_n^o \epsilon_{xy}/2 + z_n^o \epsilon_{xz}/2$$

etc., for the coordinates of the atom at the n th site. Because this strain lowers the symmetry at A and B sites, there appear lower-order terms in the crystal field. The field at an A site for example is now:

$$V_{cf}^A(u, \epsilon) = v_1^A (x'\epsilon_{yz} + y'\epsilon_{yz} + z'\epsilon_{xy}) + v_3^A x'y'z' \\ + v_2^A (z'^2 + \dots) + v_{2,1}^A (x'y'\epsilon_{xy} + \dots)$$

1(b)

Here, as in the constrained case, $x' = x - u_x$, etc. On the point charge model

$$v_1^A = \frac{3}{2} z e^2 \sum_n \frac{x_n y_n z_n}{R_n^5} a_0 \quad (2)$$

v_3^A is given by Equation (1a).

The sum is over the unstrained coordinates of near neighbors to an A site. It is clear that in general

$$\begin{aligned} v_i^A &= v_i^B, \quad i = 2n \\ v_i^A &= -v_i^B, \quad i = 2n + 1 \end{aligned} \quad (3)$$

The electron-lattice energy density, or the magneto-elastic Hamiltonian if operator equivalents are used, is the sum of contributions from each sublattice plus the elastic energy density:

$$\begin{aligned} E_e + E_{me} = & \langle V_{cf}^A \rangle + \langle V_{cf}^B \rangle + \frac{1}{2} C_{44} (\epsilon_{xy}^2 + \epsilon_{xz}^2 + \epsilon_{yz}^2) \\ & + \frac{1}{2} (C_{11} - C_{12}) (\epsilon_{xx}^2 + \epsilon_{yy}^2 + \epsilon_{zz}^2) + \frac{1}{2} k (u_x^2 + u_y^2 + u_z^2) \end{aligned} \quad (4)$$

The C's are the usual elastic moduli, k the appropriate internal modulus. Volume strains do not concern us, and have been dropped. Also, we have assumed isotropic internal forces. There are, of course, many more internal lattice modes, but they are not coupled to u_k and ϵ_{ij} , and so need not be added to the energy expression. It is more convenient to express the resulting equations in terms of the ϵ_{ij} , u_k and crystal field energy densities E_i , where $E_i = \rho V_i$. The final expression, then for the magneto-elastic energy density, suppressing terms which do not contain u_k or ϵ_{ij} or which give zero in the 4f ground state,

$$\begin{aligned} E_{me} = & 2E_1 (u_x \epsilon_{yz} + \dots) + 2E_3 (u_x \langle YZ \rangle + \dots) \\ & + 2E_{2,1} (\langle X^2 \rangle \epsilon_{xx} + \dots) + 2E_{2,2} (\langle XY \rangle \epsilon_{xy} \\ & + \dots) \end{aligned} \quad (5)$$

Actually the term in E_1 in this expression should more properly be included below in the elastic energy, since it does not depend on the angular moments of the 4f electrons; the coupling it represents persists above the Curie point and should exist even in YFe_2 , $GdFe_2$ where there are no orbital moments. It is in fact a property of all lattices of the diamond or zinc-blend structure that the internal and external strains are so coupled. Thus, a proper calculation of E_1 presents the same order of difficulty as the calculations (not attempted here) of the elastic constants and internal moduli defined below.

Finally,

$$E_e = \frac{1}{2} C_{44} (\epsilon_{xy}^2 + \epsilon_{xz}^2 + \epsilon_{yz}^2) + \frac{1}{2} (C_{11} - C_{12}) (\epsilon_{xx}^2 + \epsilon_{yy}^2 + \epsilon_{zz}^2) + \frac{1}{2} k (u_x^2 + u_y^2 + u_z^2) \quad (6)$$

The total strain-dependent energy is therefore $E = E_e + E_{me}$. The E 's all have units of energy. k is the internal modulus. We have written $E_1 = E_1^A$, etc., to simplify notation.

The Equilibrium Strains - The result of minimizing E with respect to the u 's and ϵ 's, i.e.,

$$\frac{\partial E}{\partial u_k} = 0 \quad \frac{\partial E}{\partial \epsilon_{ji}} = 0 \quad (7)$$

is

$$k u_x - 2E_1 \epsilon_{yz} = 2E_3 \langle YZ \rangle, \text{ etc.} \quad (8a)$$

$$C_{44} \epsilon_{yz} - 2E_1 u_x = 2E_{2,2} \langle YZ \rangle, \text{ etc.} \quad (8b)$$

and

$$(C_{11} - C_{12}) \epsilon_{xx} = 2E_{2,1} \langle x^2 \rangle, \text{ etc.} \quad (9)$$

Note that u_x is coupled to ϵ_{yz} , u_y to ϵ_{zx} and u_z to ϵ_{xy} . The ϵ_{ii} are not coupled to internal modes.

We deal with more-than-one electron configurations in the standard way: replace xy for example by $\sum_i x_i y_i$, where the sum is over all 4f electrons, and then using the operator-equivalent⁽⁵⁾ technique. Hence:

$$\langle XY \rangle = \alpha \langle r_f^2 \rangle (J_x J_y + J_y J_x) \quad (11)$$

α 's for various rare earths are tabulated.⁽⁵⁾ $\langle r_f^2 \rangle$ is the average of the square of the 4f electron radius, which is also tabulated.⁽⁶⁾

At temperatures well below the Curie point this expression (Equation 11) may be replaced by its zero-temperature value⁽⁷⁾

$$J_x J_y + J_y J_x \rightarrow (J^2 - \frac{J}{2}) \gamma_x \gamma_y,$$

where γ 's are the direction cosines of the magnetization.

Thus

$$\langle XY \rangle \approx J(J-1/2) \alpha \langle r_{4f}^2 \rangle \gamma_x \gamma_y \quad (12)$$

With this aside, we are now prepared to examine the expressions for the strains. Solving (8a) and (8b) for ϵ_{yz} ,

$$\epsilon_{yz} = \frac{1}{C_{44}} \left(\frac{4E_3 E_1 / k - 2E_{2,2}}{1 - 4E_1^2 / k C_{44}} \right) \langle YZ \rangle \quad (13)$$

Similar expressions hold for ϵ_{xy} and ϵ_{xz} . For the internal strain we find

$$u_x = \frac{1}{k} \left(\frac{2E_3 - 4E_1 E_{2,2} / C_{44}}{1 - 4E_1^2 / k C_{44}} \right) \langle YZ \rangle \quad (14)$$

For the lattice to be stable under these strains $kC_{44} > 4E_1^2$.

3. Numerical Estimates

We now proceed to a discussion of the sizes of the predicted effects. Although the strain modulus is known for Laves phase compounds,⁽¹⁰⁾ the internal modulus is not. In principle it could be obtained from measurements of the energy at long wavelengths of a special "optic" mode in the paramagnetic phase of these compounds. The special mode we mean is that corresponding to the two inequivalent rare earths oscillating 180° out of phase with one another while the iron sublattices sit still. It can be shown (See Appendix) rather simply that this sort of oscillation is in fact a normal mode in the cubic Laves phase if all forces are assumed isotropic. Indeed, this is the reason we could neglect any possible iron-atom displacement in the preceding discussion; such displacements just do not couple to the internal mode we are discussing. As we show in the appendix the modulus k can be written in terms of the iron-rare earth and rare earth-rare earth force constants:

$$k = 2K_{R,R} + 4K_{F,R} .$$

Although the point charge model for calculating the crystal field parameters and their strain derivatives is not expected to be accurate, we use it as a means of estimating ratios of these parameters. That is, we calculate E_3 and E_1 in terms of $E_{2,2}$ and determine $E_{2,2}$ from magnetostriction values of a metallic ferromagnet with one atom per unit cell. On the point charge model, $E_3 \approx 40E_{2,2}$ and $E_1 = -(3/4)E_{2,2}$. Assuming $4E_1^2/kC_{44} < 1$, ϵ reduces to

$$\epsilon_{yz} = \frac{2E_{2,2}}{C_{44}} (60E_{2,2}/k+1) \langle YZ \rangle .$$

To obtain an idea of the size of the internal modulus required to make the extra-ordinary term dominant in this expression, we set the ordinary part equal to a value typical of cubic ferromagnets:

$$(\epsilon_{yz})_{\text{ordinary}} = \frac{2E_{2,2}}{C_{44}} \langle YZ \rangle \sim 10^{-5} .$$

(10) Clark, A. E. and Savage H. T., IEEE Trans. Sonics and Ultrasonics SU-22, 50, January 1975.

From tables⁽⁸⁾ of Steven's factors we find that, for Tb⁽⁹⁾ at low temperatures,

$$\langle YZ \rangle = .33 \times 10^{-3}$$

Here we have used $r_f^2 = .185 \text{ \AA}^2$ and $a_0 = 8 \text{ \AA}$. (The radius was obtained from reference (6) and the lattice constant from reference (1).) Finally, using

$$C_{44} = 2 \times 10^{11} \text{ ergs/cm}^3$$

we get

$$E_{2,2} = 10^9 \text{ ergs/cm}^3$$

Thus, for the "extra-ordinary" part of the strain to dominate, and give the experimental result for TbFe₂, we require $E_{2,2}/k \approx 1$, which means the normal frequency of the internal mode in the paramagnetic phase would have to be 10^{12} cps or lower. Such frequencies are typical of optic modes in metals with more than one atom per cell. In sum, the size and anisotropy of the magnetostriction in Laves phase magnets can be accounted for on the basis of the theory given here, culminating in Equation (13). The magnetostriction is predicted to be positive for TbFe₂, (α is negative) on the basis of Equation (12) if the extraordinary term dominates. This accords with experiment.⁽²⁾

The prediction of an internal distortion can be checked by X-ray measurements, though the analysis is complicated by the distortion caused by the external strain. We want to make a remark about the direction of the internal distortion. It follows from Equation (14) that the distortion is parallel to the magnetization when the latter lies along a [111] axis; it vanishes when the moment is parallel [100]. When the moment is in the xy plane, the A and B sublattices are alternately distorted positively and negatively along the Z axis. For a general direction of magnetization the two sublattices undergo antiparallel distortions but the axis of the internal distortion is not along the moment direction whereas external distortion is along magnetization direction. This strange behavior is consistent with

(8) Freeman, A. J. and Watson, R. E., Phys. Rev. 127, 2058 (1962).

(9) Tsuya, N. Clark, A. E., and Bozorth, R., Proc. International Conf. on Magnetism, Nottingham, p. 250 (1964).

the physical picture we gave in Section 1: The 4f charge cloud always tries to move so as to get closer to its nearest set of neighbors (See Figure 2).

Finally, we note that these effects should also occur in the isostructural Co and Ni Cl₅ compounds with the rare earths at temperatures below their respective transition temperatures. A similar internal distortion will also take place in the magnetic phases of the similar Cl₄ compounds, with the added complication that the latter are hexagonal, so that there are more than two external strain modes.

In our description of the atomic displacements we have ignored distortions of the transition metal atoms and taken the rare earths to move equal amounts in opposite directions. For this theory to be a complete account of the magnetostriction requires that this rare earth movement be a normal mode of vibration in the absence of strain and that there be no other modes, i.e., iron sublattice motion, which couple to the external strain. The first of these requirements is satisfied, as we show in the Appendix. The second is not, a situation we will deal with in a subsequent publication. The rare earth displacement is, we feel, the most important one for considerations of magnetostriction.

APPENDIX A

Our purpose here is to show that we are justified in assuming that $u_A = -u_B$, i.e., that is no coupling to distortions of the transition metal sublattices. (That all the rare earth atoms of a given sublattice undergo the same distortion follows from translational invariance.) We do this by calculating the normal modes of the cubic Laves phase assuming near-neighbor coupling. For the set of solutions which vary in time like $\exp(-i\omega t)$ in the limit of long wavelength the equations of motion are

$$-\omega^2 M_\alpha u_\alpha^i = \sum_j \sum_{\beta \neq \alpha} K_{\alpha,\beta}^{ij} (u_\beta^i - u_\alpha^j).$$

Where α and β are sublattice indices running from one to six, and i and j refer to x , y , or z components.

The force constants $K_{\alpha\beta}^{ij}$ are defined as:

$$K_{\alpha\beta}^{ij} = \nabla_i \nabla_j \sum_n V(R_n),$$

where the sum is over all near neighbors to an α site which are on the β sublattice. Label the rare earth atoms by $\alpha = 5$ and 6 . If V depends only on the distance between sites then $K_{\alpha\beta}^{ij} = 0$ if $i \neq j$ and $\alpha = 5, \beta = 6$. Also $K_{5,\beta}^{ij} = K_{6,\beta}^{ij}$ for $\beta = 1$ through 4 . Then

$$\begin{aligned} -\omega^2 M u_5^i &= K_{5,\beta}^{ii} (u_\beta^i - u_5^i) + \sum_j \sum_{\beta=1}^4 K_{5,\beta}^{ij} (u_\beta^j - u_5^j) \\ -\omega^2 M u_6^i &= K_{6,\beta}^{ii} (u_\beta^i - u_6^i) + \sum_j \sum_{\beta=1}^4 K_{5,\beta}^{ij} (u_\beta^i - u_6^i). \end{aligned}$$

Here M is the rare earth mass. It can also be shown from the definition of $K_{\alpha\beta}^{ii}$ that $K_{5,\beta}^{ii} = K_{6,\beta}^{ii}$ so that, by subtracting the last two equations

$$-\omega^2 M (u_5^i - u_6^i) = 2K_{5,6} (u_6^i - u_5^i) - (u_5^i - u_6^i) \sum_{\beta=1}^4 K_{5,\beta} .$$

Defining $K_{5,6} = K_{R,R}$ and $\sum_{\beta=1}^4 K_{5,\beta} = 4K_{F,R}$ we see that there exists a normal mode in which the two rare earths beat against one another with a frequency

$$\omega = \sqrt{\frac{2K_{R,R} + 4K_{F,R}}{M}}$$

Further, the normal coordinates for this mode have the property that $u_A = -u_B$, $u_\beta = 0$ for all four transition metal atoms. This is exactly the form of static distortion we have assumed in Section 2.

Figure Captions

Figure 1

A drawing of the atomic arrangements in the C15 structure. The R atoms are arranged identically to the carbon atoms in the diamond lattice. Each unit cell has two R (rare earth) and four T (transition metal) sites.

Figure 2

The distortion along [111] which results from the different net Coulomb attractions of the two rare earth ions in the C15 unit cell.

Figure 3

When [100] is easy, there is no distortion; the charge cloud on the rare earth remains equidistant from all its neighbors.

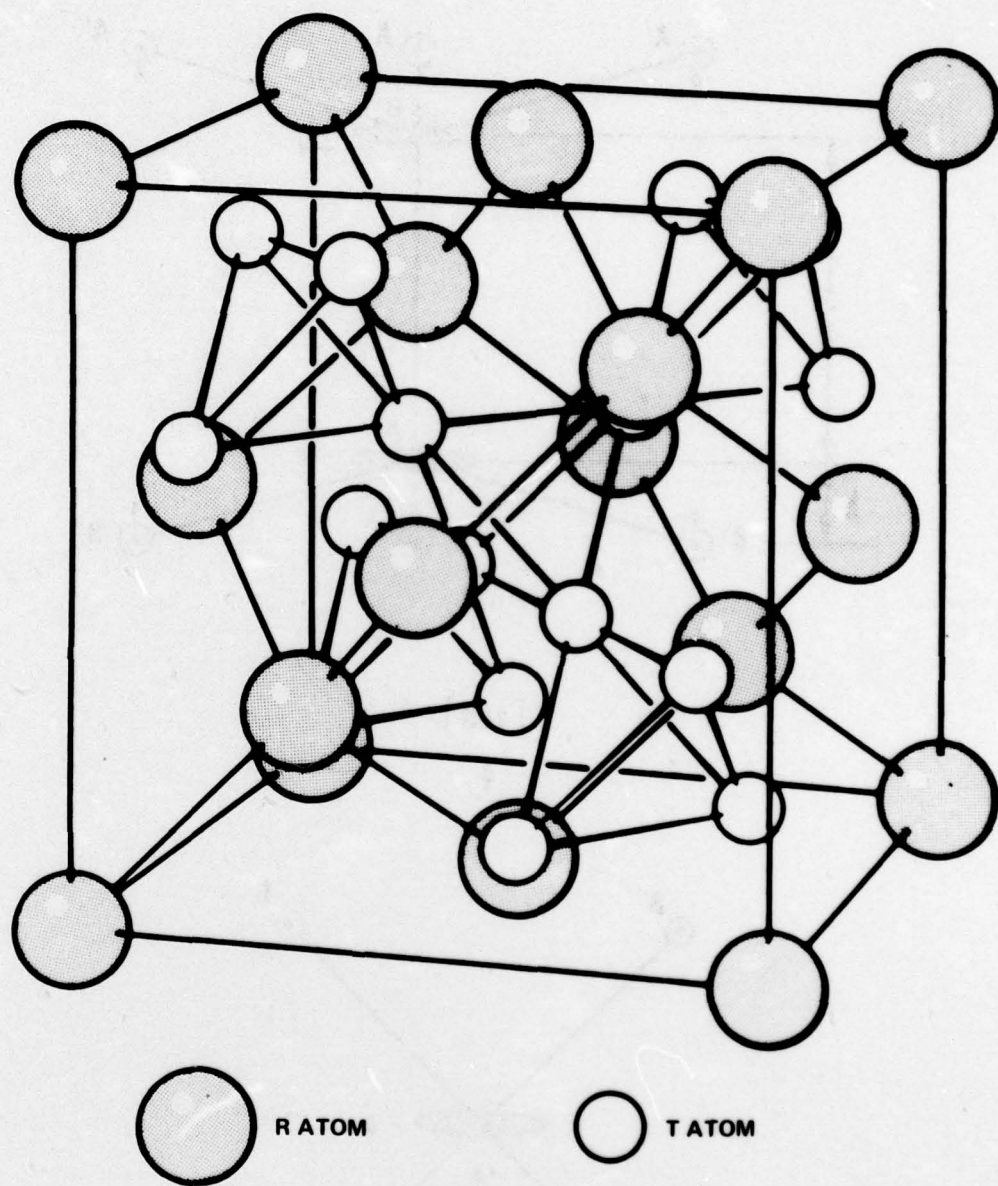
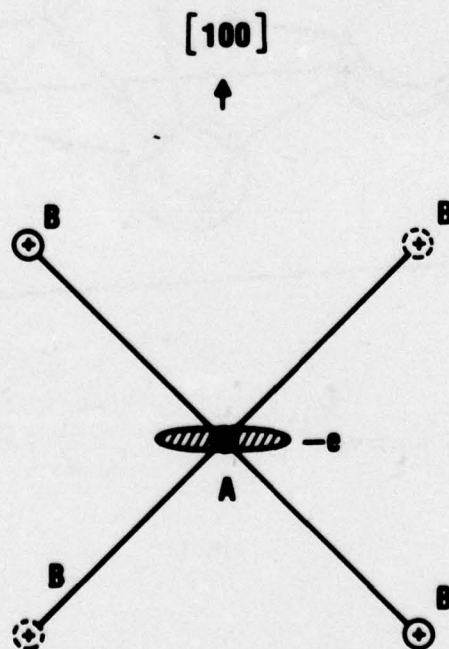
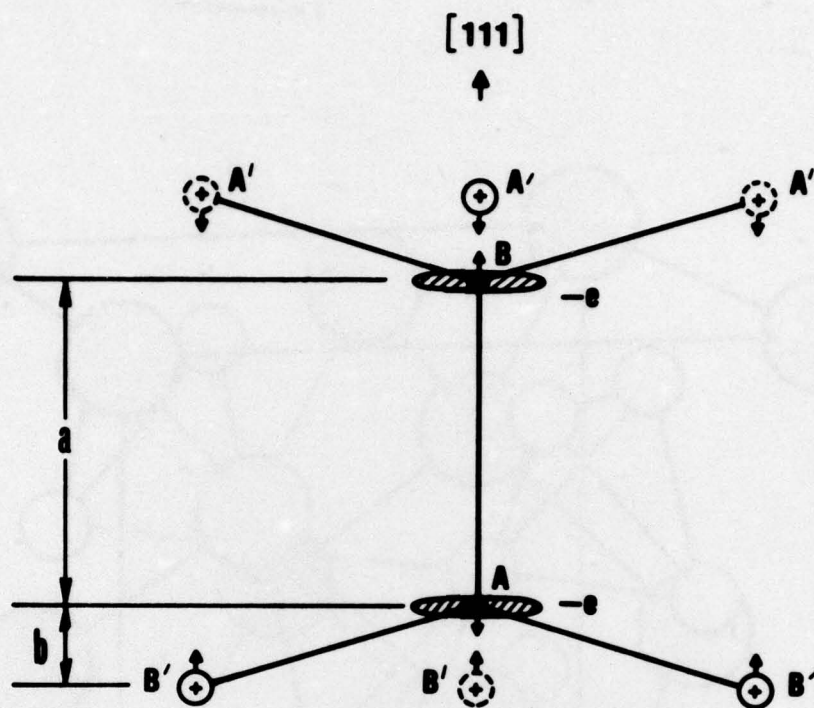


FIG. 1



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